



A note of relationship between outdoor and indoor exposure integrals for air pollution of outdoor origin

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Publication date:
1980

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Gjørup, H. L., & Roed, J. (1980). *A note of relationship between outdoor and indoor exposure integrals for air pollution of outdoor origin*. Risø National Laboratory. Risø-M No. 2234

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RISØ-M-2234

A NOTE ON THE RELATIONSHIP BETWEEN OUTDOOR AND INDOOR
EXPOSURE INTEGRALS FOR AIR POLLUTION OF OUTDOOR ORIGIN

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Abstract. Beryllium-7 created by cosmic radiation has been used as a tracer in preliminary measurements designed to enable an estimation of the ratio between outdoor and indoor exposure integrals for aerosols of outdoor origin, with special reference to the reduction in inhalation dose that can be achieved by staying indoors during reactor accidents. Earlier investigations relevant to this problem are reviewed.

It is concluded that the reduction in inhalation dose offered by an average Danish house is roughly one order of magnitude.

INIS-descriptors: AIR POLLUTION, BERYLLIUM 7, BUILDINGS, INHALATION, IODINE 131, MEASURING METHODS, RADIATION DOSES, RADIOACTIVE AEROSOLS, REACTOR ACCIDENTS, REVIEWS, TRACER TECHNIQUES.

UDC 614.73 : 628.8 : 621.039,58

May 1980

Risø National Laboratory, DK 4000 Roskilde, Denmark

ISBN 87-550-0676-0

ISSN 0418-6435

Risø repro 1980

CONTENTS

	Page
1. INTRODUCTION	5
2. GENERAL INFORMATION	5
3. SURVEY OF RELEVANT STUDIES AVAILABLE	6
4. THE WINDSCALE STUDY	8
5. THE PITTSBURGH STUDY	11
6. BERYLLIUM-7 INVESTIGATIONS.....	12
7. DISCUSSION	14
8. CONCLUSION	16
REFERENCES	17

1. INTRODUCTION

The object of this note is to estimate to what degree the filtering effect of cracks, crevices and pores, as well as any possible deposition on floors, walls and furnishings, can reduce inhalation doses on Danish territory to people remaining inside buildings during a large, hypothetical reactor accident at the Barsebäck nuclear power station - such as is described, for example, in the report Rise-M-1905. The present note is a translation from its counterpart in Danish dated May 1979.

2. GENERAL INFORMATION

Any pollution of inhaled air means that the polluting material will be taken up by the lungs in an amount that is proportional to the inhalation rate and to the exposure integral. The exposure integral is the integral of the concentration in the air as a function of time.

People inside buildings will normally obtain some protection against air pollution of outside origin. This is the result of physical and chemical processes. Firstly, some of the pollution will be removed by filtration in the cracks, crevices and pores through which air penetrates into buildings. Thereby, the pollution concentration will be less indoors than outdoors. Secondly, some of the pollution that penetrates into the inside may be deposited on floors, walls, ceilings and furniture. This will contribute to reduce the concentration in the indoor air. Finally, conversion processes, such as radioactive decay, may also contribute to reduce the concentration.

When dealing with a relatively short-term air pollution episode - such as a reactor accident - it is possible to achieve a further reduction of the indoor exposure integral, and thus of the inhalation doses, by deliberately controlling ventilation

in rooms (e.g., by closing and opening windows). As long as the air exchange does not alter, it in itself, regardless of its magnitude, will not affect the exposure integral, i.e., apart from the influence that it may have on some of the factors mentioned above. Disregarding these factors, the total exposure integral resulting from the passage of a pollution cloud will be the same, indoors and outdoors, provided that the air exchange remains constant. Even though the concentration builds up more slowly indoors than outdoors, it will, on the other hand decrease more slowly too. In this manner the same total air pollution is simply made to be of longer duration indoors than outdoors. This means, however, also that the exposure integral indoors can be reduced by deliberately increasing the air exchange (e.g., by opening the windows) after the pollution cloud has passed by. In contrast, of course, the exposure integral can be increased by reducing the air exchange (e.g., by closing any windows that may be open) when the cloud has passed. The degree of reduction that can be achieved in this manner depends on the natural air exchange in the building in question. The less the natural air exchange, the greater the reduction. As indicated earlier, air exchange is also of importance for the effectiveness of a deposition of polluting materials on various surfaces inside a building. Furthermore, it must be assumed that there is some connection between the natural air exchange and the effectiveness of the filtering effect.

3. SURVEY OF RELEVANT STUDIES AVAILABLE

In spite of the fact that the circumstances mentioned above - in particular the filtering effect - may be expected to have a considerable influence on the hazard to man implied by air pollution, it seems, at least as far as the present authors are able to ascertain, that only two relevant experimental studies (Megaw, 1961 and Alzona et al., 1979) have been carried out, prior to our own preliminary Beryllium-7 measurements (reported in this note) on the magnitude of the reduction that may be achieved.

Both of the investigations in question had the same specific aim, that we have, namely a desire for a realistic evaluation of inhalation doses during reactor accidents.

The first investigation was carried out by Jim Megaw after the Windscale accident in 1957 - this will be called the Windscale study in the following. The other investigation was carried out by J. Alzona et al. at Pittsburgh University in 1978 - this will be called the Pittsburgh study.

At the Health Physics Department of the Risø National Laboratory we have carried out preliminary investigations of the relationship between outdoor and indoor exposure integrals. The measurements carried out so far, which are discussed in the following, are based on a new method that utilizes the radioactive isotope ^7Be as tracer. Beryllium-7 is chiefly created in the stratosphere by spallation processes resulting from cosmic radiation. Even though a small amount of ^7Be is created on the ground, and hence also indoors, this material can be considered for all practical purposes as a pollutant of exclusively outdoor origin because its half-life is very long (about 53 days) compared with the time constant for air exchange in houses (which is measured in hours).

The Windscale and Pittsburgh studies are more closely discussed in the following, where they are compared with Risø's preliminary investigations. The two studies are rather incomplete and Risø's investigations are only the first steps of work started in 1978 which it is planned to continue in 1979. However, on the basis of these investigations, and on the measurements of air exchange in Danish houses carried out by the (Danish) Technological Institute (Collet et al., 1976), it seems possible to estimate the size of the protection factor with reasonable accuracy. Presumably the continued investigations will be able to improve upon this accuracy.

4. THE WINDSCALE STUDY

The reactor accident at Windscale in October 1957 offered a unique opportunity of studying the protective effect of buildings, as it involved a radioactive aerosol that, due to its very nature, possessed the characteristics expected in a reactor accident, and which was sufficiently concentrated to make it possible to make accurate measurements, also at larger distances.

One week after the accident the deposition of ^{131}I was measured indoors and outdoors in an office building at Drigg, located about 6 km from Windscale, and in a two-storey stone-built house at High Saltcoats, located 9 km from Windscale. Both buildings lay in the direction of the smoke plume, and neither was in use during the period of time in question. The house in High Saltcoats had sash windows and six chimneys. Some of the windows in the office building at Drigg were open during the accident. Because of the workload on the technical staff in connection with the accident, measurements were only started a week after the incident and it was thus not possible to measure the indoor and outdoor exposure integrals such as would have been desirable. For this reason the Drigg and Saltcoats measurements were later supplemented by an experiment carried out at the old Harwell airbase. Here there was a wooden hut, described as relatively newly erected, well maintained and with tight-fitting windows and reasonably tight-fitting doors. This experiment involved the release of radioactive iodine at a distance of approximately 20 m from the hut. Measurements were then made of the deposition and the indoor and outdoor exposure integrals. In addition, the air exchange in the hut was measured before each series of experiments. Finally, the exposure integrals for an inactive aerosol composed of Aitken nuclei were also measured.

The deposition of iodine on pieces of paper left in the office building at Drigg was about 0.7% of the deposition on surfaces outside the building. There were no pieces of paper available in

the house at High Saltcoats, which could easily be measured, but comparative measurements were made of the iodine content of the dust on the floors at Drigg and High Saltcoats. This comparison showed that the deposition indoors at High Saltcoats was only about 0.16 times the deposition indoors at Drigg, corrected for the relationship between the outdoor depositions and for the weight of dust per m^2 . That is, the deposition in the house at High Saltcoats was only about 1 o/oo or 1/1 000 of the outdoor deposition, if paper can be assumed representative of the surfaces in a house.

Taking the measured relation between the deposition velocities^{x)} for iodine outdoors and indoors in the hut experiment at Harwell, which was about a factor 20, as generally valid, it could be concluded that the indoor exposure integral at High Saltcoats was about $\frac{1}{1000} \times 20 = \frac{1}{50}$ of the outdoor exposure integral, or that the house gave a protection factor of about 50 for inhalation doses. If the indoor deposition velocity was about 0.1 cm/sec, as measured at Harwell, the above conclusion does not seem unreasonable since such a great deposition velocity implies that a large part of the incoming iodine would be deposited on surfaces inside the house, whereby the exposure integral would be significantly reduced - perhaps by a factor 10 - the remaining factor 5 being explained by filtration.

Nevertheless, the conclusion can hardly be correct. Recalling that some of the windows in the office building at Drigg were open during the accident, it is reasonable to assume that the

^{x)} The deposition velocity is defined as

$$\frac{\text{total deposition/cm}^2}{\text{exposure integral secs/cm}^3}$$

indoor exposure integral was just as large or nearly as large as the corresponding outdoor exposure integral. Yet, the indoor deposition was measured to about 0.7% of the outdoor deposition. This means that the deposition velocity, too, was 0.7%, or perhaps a little more, of the outdoor deposition velocity, i.e. about 0.007 cm/sec, which is at variance with the result of the Harwell measurement that showed 0.1 cm/sec indoors.

A deposition velocity as low as 0.007 cm/sec will not affect the exposure integral appreciably in a house with as large an air exchange as the one in High Saltcoats is assumed to have had from its description (unfortunately the air exchange was not measured, but doubtless it was large compared with average Danish houses). Furthermore, it is reasonable to assume that the deposition velocity for iodine on floor dust at Drigg and at High Saltcoats was the same (Sehmel, 1973); The above reasoning leads inevitably to the conclusion that the house at High Saltcoats afforded protection, with respect to the inhalation dose for iodine-131, of a factor about 6, or perhaps somewhat more, and that this protection factor resulted exclusively from filtering.

Hence it must be inferred that the Harwell experiments using radioactive iodine are without relevance in this connection - at any rate with respect to the deposition velocity. It should be recalled that the iodine source used was placed at a distance of 20 m from the hut; therefore the aerosol can hardly be assumed to be representative of an aerosol produced at a distance of 9 km. Furthermore, copper disks were used for the measurements of deposition, and this material is hardly representative of the surfaces in a house, in particular not with respect to free iodine.

The results of the measurements in the Harwell experiments may otherwise be described briefly as follows. The relationship between the outdoor and indoor exposure integrals for iodine was about 3, taking the average of five measurements, and the average air exchange was about three times an hour. The relation

between the outdoor and indoor exposure integrals for Aitken nuclei (produced outside the hut, particle size from a few hundredths of a micrometer and upwards) was about 1.7 for the average of five measurements, and the accompanying average air exchange was about 1.7 times an hour.

5. THE PITTSBURGH STUDY

The purpose of this study was to evaluate the protection factor against inhalation of dust of outdoor origin for people being indoors. The method consisted of taking filter samples of air and determining the content of collected particles of calcium, iron, zinc, lead and bromine by means of X-ray fluorescence analysis.

The particles in question are known to be principally of outdoor origin. The particle sizes were not measured in connection with the study, but reference is made to other investigations, where they were found typically to be the following: calcium 0.65-20 micrometer; iron 3.6-20 micrometer; zinc 0.65-29 micrometer; lead 0.1-0.65 micrometer; bromine 0.1-0.65 micrometer. The lead and bromine associated with particles originate chiefly from automobile exhaust gas, and the concentration varies widely during the course of a day. The iron and zinc levels in Pittsburgh are much higher than in other areas, which shows that they are principally of industrial origin. Calcium is an important component of fly ash, and it is also released when using limestone in the production of steel.

The ratio between outdoor and indoor concentrations in equilibrium was measured in ten rooms of widely differing character (two of them were cars). The greatest value for this ratio was found in a 10 m² room without windows in a new university building in Pittsburgh, and it is stated to have been greater than 10 for all types of pollution measured (iron, lead and bromine). For the other nine rooms, the average for bromine was 2.79; for

lead 2.38; for iron 4.17; for zinc (only five rooms) 2.44; and for calcium (only four rooms) 10. The lowest value - 1.43 - was found in a 30 m² large bedroom with eight windows.

One of the rooms - a 12 m² office in an old university building with a large six-paned window, of which two sections could be opened - was investigated in greater detail. It was measured as it was (case J), with plastic foil over the window (case K), with plastic foil covering all surfaces (case N), and with plastic foil covering all surfaces except the window (case P). These experiments seem to show that the outdoor pollution exclusively, or almost exclusively, penetrated through the window. In case J the relation between the outdoor and the indoor exposure was 10 for calcium, 5.88 for iron, 1.92 for zinc, 2.04 for lead, and 2.78 for bromine. The experimenters corrected these values for background concentrations indoors on the basis of case N, and thereby found a protection factor of between 3 and 20, but they also point out that the protection factor becomes much greater for calcium, iron and zinc if correction is made on the basis of case K. Relating to case K does not significantly alter the factor for lead and for bromine, which was 2.94 and 7.69, respectively.

Naturally, these figures only apply to the room investigated and the correction procedure appears doubtful. The study would have been considerably more valuable had the air exchange been measured.

6. BERYLLIUM-7 INVESTIGATIONS

As already mentioned, ⁷Be is created in the stratosphere and the concentration at ground level is very low, especially in the autumn and winter. The outdoor concentration has varied from about 40 to about 170 picocuries per 1000 m³ in the measuring periods during the course of the autumn and winter. The collecting of filter samples will further reduce the indoor

exposure integral perceptibly if the suction speed is too great. The samples are measured with a very efficient gamma spectrometer; in order to achieve sufficiently good measuring accuracy, however, it is nevertheless necessary to stretch the sample collecting period over one to two weeks. During this time the houses concerned cannot be used because windows and doors must be kept closed. Therefore objects for measuring were very limited in the introductory phase of the study; they consisted of a former grocer's shop and a farmhouse, both buildings now being used as summer homes (called A and B, respectively, in the following), and furthermore a more than 20-year-old, poorly maintained single-family house (called C in the following).

In house A the ratio between the outdoor and indoor exposure integrals in a room opening onto the garden was about 7, taking the average of five measurements. The highest measured value was about 9 and the lowest about 4.5. The doors were closed in the whole house during the sample collecting period. Air exchange in the room in question was measured to about 1.0 times an hour. Furthermore, two measurements were made in the dining-room, one with closed doors and the other with open doors in the whole house. The exposure integral ratios were measured to about 2.6 and 1.9, respectively. The corresponding air exchange values in the room concerned were, respectively, about 3.5 and 4.5 times per hour.

In house B the ratio between the outdoor and the indoor exposure integrals in the kitchen was about 2.0, as an average of four measurements. The highest value was about 2.4 and the lowest 1.7 (measured during a snowstorm at the turn of the year). The doors were kept closed in the whole house during the sample collecting periods. The air exchange in the room in question was measured to about 4.5 times an hour.

In house C only one measurement was made of the exposure integral ratio. It was about 6 in the living room with doors closed in the whole house. The air exchange was measured to roughly once an hour in the room in question.

In addition, a measurement was made of the deposition of ^7Be indoors in house C. At first, the whole of the floor in the living room was covered with plastic foil for a week. The plastic foil was then compacted and measured, but no activity was detectable on it. The floor, which had been polished previously, was then planed off and the planing dust was pressed into pellets. It proved possible to measure the ^7Be activity in these pellets, corresponding to a deposition velocity of about 0.004 cm/sec. This means that indoor deposition is of no practical significance.

It is obvious that further houses should be included in the continued studies, and especially those with less air exchange, so that the full spectrum of Danish housing can be covered with respect to air exchange. Moreover, an attempt should be made to determine the particle spectrum of ^7Be . In the further studies we also intend to use X-ray fluorescence analyses and the anthropogenous lead pollution of air as trace substance, i.e., in principle the same technique as used in the Pittsburgh study.

7. DISCUSSION

The most important single factor ascertained in this note is that a two-storey house having sash windows and six chimneys (and presumably a corresponding number of open fireplaces), located about 9 km from Windscale during the accident that occurred at this station in October 1957, can be assumed with great certainty to have provided protection against the inhalation of ^{131}I of a factor 6, or perhaps somewhat more. The observation is of special importance because this is the only case where there has been an opportunity to study conditions relating to an aerosol produced by a reactor accident, and to do so at a large distance from the source point. The observation is of further importance because it concerns iodine, which would be one of the most important isotopes connected with both

larger and smaller reactor accidents resulting in the release of fission products to the atmosphere.

The Windscale study suffers from the deficiency that the air exchange was not measured in the house in question, but according to the description of this house the air exchange was presumably much greater than that of the average Danish home.

As a first approximation it seems reasonable to assume that the effectiveness of the filtration is inversely proportional to the air exchange in the different types of building. This assumption is confirmed by the ^7Be measurements for large and medium air exchange values. Extrapolating from these measurements to the mean value of air exchange for Danish houses (about 0.63 times an hour), as was measured by the Technological Institute (Collet et al., 1976) a filter factor of about 10 is found.

On the other hand, the Pittsburgh study shows protection factors as low as about 3-7 for lead and bromine, where the particle size is stated to lie between 0.1 and 0.65 micrometer (corresponding to the size that can presumably be expected at some km distance from a source point (Schjoldager, 1978)). These figures are, however, based on measurements in a single room, where the air exchange is unknown, and on uncertain corrections. Hence no great importance can be attached to them.

All in all it must be assumed reasonable to ascribe to the estimated protection factor of about 10 for the average Danish House an uncertainty of a factor 2, both upwards and downwards. The scatter around the mean value for all Danish houses is estimated, on the basis of the measurements made by the Technological Institute, to be a factor about 2.2, (corresponding to 2 standard deviations assuming a logarithmic distribution).

It should be noted that the measurements carried out by the Technological Institute of the air exchange (Collet et al., 1976) were performed with inner doors open; the air exchange might be less with inner doors closed. A special study made by this Institute at the request of the Ministry of the

Environment, which comprised three houses, showed only an effect of this kind in the case of the house with the greatest air exchange. For the other houses, there seemed to be no air exchange whatsoever between the individual rooms when the inner doors were kept closed, and at the same time the total air exchange in these two houses was only affected to a slight degree. This could be an accidental result and it is desirable to carry out further measurements with inner doors closed. It is also desirable to investigate to what extent air exchange can be reduced by simple insulation measures.

All the figures given above for protection factors relate to the filtering effect only.

8. CONCLUSION

On the basis of the present very limited material, it is estimated that the inhalation doses from an aerosol produced by a reactor accident at some km distance from the source point, can be reduced by a factor of about 10 for people remaining inside the average Danish house because of the filtering effect alone, if one compares with what the inhalation doses would have been outside the house.

The reduction factor in question must be considered to be encumbered with an uncertainty factor of about 2, both in an upward and a downward direction. The scatter around the mean value is estimated to be a factor of about 2.2 for all Danish houses (it is 2 standard deviations assuming a logarithmic distribution). However, there are strong indications that the reduction factor for iodine is no less than a factor about 6, even for houses with very large air exchange.

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Date 1980-06-16

Department or group

Health Physics

Group's own registration
number(s)

TM 269

pages + tables + illustrations

Abstract

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